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COMPARISON OF MOLECULAR DYNAMICS CALCULATIONS
WITH OBSERVED INITIATION PHENOMENA*

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SUMMARY

We have completed, over the past three years, a large number of molecular dynamics calculations of the effects of the interaction of shock waves with condensed systems representing chemical explosives. Our capabilities have recently been expanded to permit 3-D calculations of quite realistic organic explosives systems and to provide study of the initial chemical interactions by keeping current the spatial relationships of each of the atoms in a specified system with relation to all the others.

We have explored the microscopic effects of material characteristics known to cause changes in the sensitivity of explosives -- voids and cracks, crystal defects, inclusions, initial temperature, and shock strength, and we have found a very instructive correlation with the macroscopic results. We would present a film summary of these calculations.

The technique of molecular dynamics has evolved to a more sophisticated level because of the dramatic revolution in large-scale computer technology. Simply stated, molecular dynamics involves the numerical solution by computer of Newton's equations of motion for all the atoms comprising the active region of the assembly. As a result the coordinates and velocities of the particles are obtained as functions of time.

The set of coupled nonlinear, second-order differential equations that represent the forces are reduced to a doubled set of first-order ordinary differential equations by standard procedures. The initial positions and velocities of the particles represent the initial conditions on these equations. For the numerical solution of this set of equations we have employed the variable-step, variable-order, implicit Adams method with functional (or fixed point) iteration; a generalization of the classical Adams-Bachforth-Moulton method. The force acting on any one particle in our ensemble is the resultant of all interactions with other atoms in its neighborhood, and it is obtained as the derivative of an effective many-body potential. The correctness of our assumed potentials is indicated by the agreement with experiment obtained in calculation of the appropriate shock "Hugoniots." Representative potentials we are using are presented in Fig. 1.

Extensive studies of voids in model two-dimensional diatomic systems, such as face-centered cubic nitric oxide and sulphur nitride lattices impacted by aluminum plates, have been carried out. Fig. 2 is representative of such studies and is used for illustration. We find the qualitative behavior of all such systems to be much the same, whether the system is initially quiescent or thermal. The shock, upon reaching the first wall of the void, spalls atomic fragments into it, which are moving in a highly non-random, non-ergodic fashion and traverse the void to produce very localized and specific bond rupture in the opposite wall. Again, this is shown dramatically in the later events of Fig. 2.

There is good experimental evidence that chemical bonds can be broken by strictly mechanical forces to form free radicals, and our calculations indicate that a similar process can occur in a condensed explosive material within a shock front. These calculations indicate that this process is not in thermal equilibrium nor does it obey the rules of statistical mechanics. It appears to take place in 10^{-13} to 10^{-10} sec. (The shock wave passes over a typical explosives bond in a period that represents about one vibration). The energy of the shock appears to be very coherent and, in a homogeneous system, to have soliton-like behavior.

In studies of initiation in which we use the "reactive" potentials, the energy release is found to be highly non-ergodic, with the energy imparted to the lattice not readily degrading into random thermal motion but staying in the shock front until the shock encounters a surface. We find that the lattice at later stages is consumed by an ever-growing sequence of spalls occurring on opposite faces to the right and left of the lattice, both for the initially quiescent case and for a lattice with thermal motion initially equivalent to room temperature. The left-hand spalling sequence is found to be initiated by the arrival at the left edge of the backward-traveling disturbances generated by the right-hand spalling. The manner of this rapid destruction is such that at no stage is the system ergodic and undergoing random thermal decomposition; and there is thus no possibility of describing it by some type of simple Arrhenius rate law.

A fundamental premise of pursuing the establishment of this model is that if the initial and transitional stages of the processes involved (both the chemistry and physics) are understood, the probability for affecting those same processes is enhanced.

Shock-induced fast-decomposition processes are, of course, but one of a wide variety of phenomena brought about by shock passage through condensed matter and are the subject of extensive investigation today, e.g., such phenomena also include high-pressure polymorphic phase transitions, shock-induced polymerization, and shock-induced materials syntheses, and they are being extensively explored, particularly in the Soviet Union.

Inert

$$V(r) = D_e \left[e^{-2\beta(r - r_0)} - 2e^{-\beta(r - r_0)} \right]$$

"Reactive"
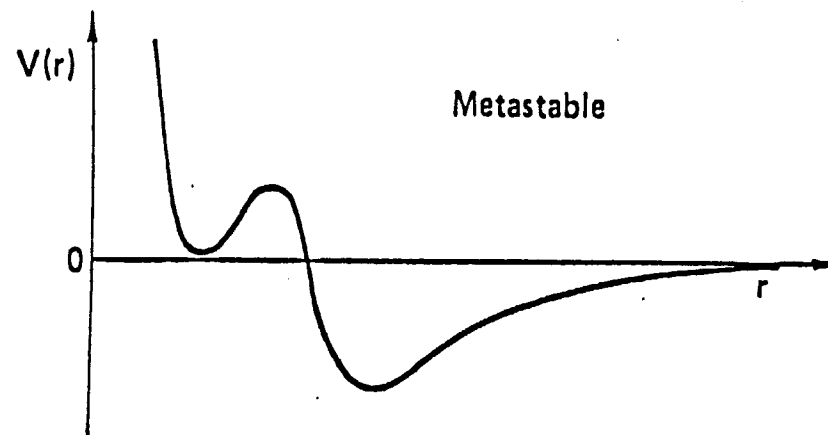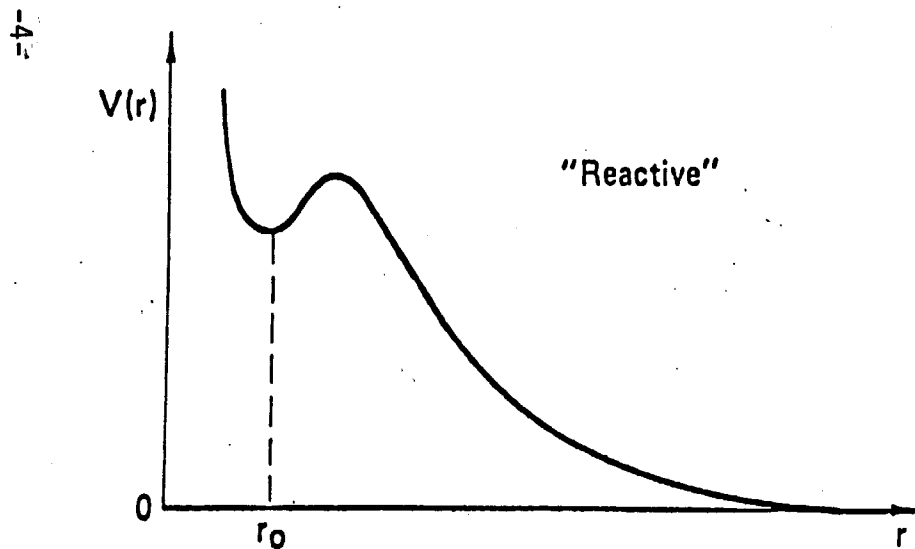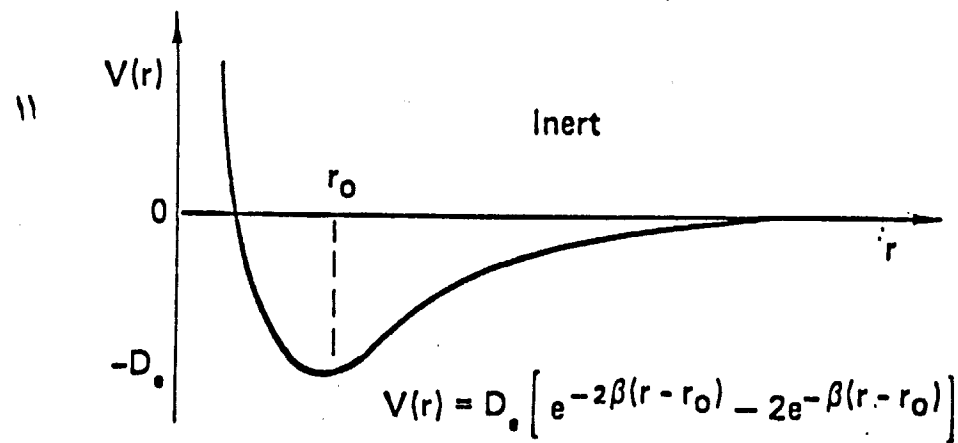
Metastable

$$V(r) = \sum_n a_n (r - r_0)^n e^{-\rho(r - r_0)}$$

Fig. 1 - Representative interatomic potentials for Morse ("inert"), "reactive," and metastable lattices.

Fig. 2

(a) t = 6

(b) t = 10

(c) t = 18

(d) t = 25

(e) t = 36

(f) t = 100

Distance — natural units

Distance — natural units